The Backscattering Enigma in Natural Waters

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LONG TERM GOALS

One of the fundamental problems in ocean optics over the past several decades has been a lack of understanding of the source of backscattering in the ocean. Because of experimental limitations and the limitations in available theoretical models, our knowledge of the causative agents for backscattering remains poor. Experimentally, we have been limited by a lack of scattering sensor instrumentation and a methodology for routine measurement of the submicron particle size distribution. Theoretically, most models have used Mie theory with the hope that natural particles of complex shape and structure can be approximated well by homogeneous spheres. For the Navy, poorly parameterized backscattering compromises applications involving the interpretation of passive and active optical detection methods. This is particularly true in coastal regions where current inversion models fail because the effects of changing particle composition are not adequately understood. Our long term goal is to better understand the sources and distribution of backscattering in natural waters.

OBJECTIVE

The apparent enigma of oceanic backscattering lies in the fact that available experimental and theoretical results do not agree. Stramski and Kiefer (1991) modeled the backscattering of representative populations of microorganisms in the ocean with Mie theory and concluded that microorganisms could only account for ~20% of the backscattering necessary to explain the upwelling radiance observed by satellites and in situ measurements. Our objective is to evaluate three hypotheses that have emerged to explain the "missing backscattering."

We call the first hypothesis the *detrital hypothesis*. Stramski and Kiefer (1991) postulated that this missing agent is high concentrations of submicron detrital particles with high refractive index. Furthermore, the authors postulated that these detrital particles should covary with chlorophyll containing particles to satisfactorily explain the observation that satellite ocean color measurements are reasonably successful in determining chlorophyll concentrations from upwelled radiance.
We call the second hypothesis the *complex particle hypothesis*. Zaneveld et al. (1974), Kitchen and Zaneveld (1992), and Zaneveld and Kitchen (1995) asserted that the bulk refractive index (and the relative amount of backscattering) for phytoplankton would be higher than that typically used in

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models based on Mie theory that assume particle homogeneity because these organisms usually, particularly in coastal regions, have a hard shell (silicious frustule in the case of diatoms). It is also possible that the scattering properties of phytoplankton particles are poorly modeled as perfect spheres. Thus, the hypothesis here is that the "missing backscattering" is due, at least in part, to deficiencies in modeling efforts.

We call the third hypothesis the *mineral hypothesis*. Twardowski et al. (2001) suggested that the unexplained backscattering could result from a background population of small inorganic particles. These particles are present throughout the ocean, primarily from biogenic and Aeolian sources, and exhibit strong relative backscatter due to their high refractive index. In this case, the "missing backscattering" is due to an inorganic mineral background, with a fluctuating phytoplankton population superimposed on that background.

APPROACH

Testing the complex particle hypothesis

A key focus over the last year has been determining the scattering properties of phytoplankton populations and understanding how best to model these properties in a bulk sense. Once we have reasonable success with the forward problem, we will then be able to attempt to carry out the inverse problem. Work has been carried out in the laboratory on various cultures of phytoplankton. Field data has also been used to test this hypothesis under conditions where phytoplankton specific scattering data may be reasonably approximated. Phytoplankton for culturing are being isolated from Narragansett Bay and surrounding waters to avoid using perpetually cultured organisms that can develop morphological inconsistencies with their naturally occurring cousins. Table 1 lists the measurements being made.

Table 1. Measurements to test backscattering hypotheses.

Physical parameter	Instrument	Specific
Volume Scattering Function, β (m^{-1} sr^{-1})	WET Labs ECO sensors	β_p ([100°, 117°, 125°, 150°], [450, 532, 650 nm])
	Goniometer	β_p ([~15°-155° @ 1 resolution], [532 nm])
Attenuation, absorption, scattering coefficients (m^{-1})	WET Labs AC9 and ACS instruments	a_{pg}, a_g, a_p, b_p, c_{pg}, and c_p at 84 wavelengths
Particle Size Distributions (PSDs)	LISST-100	Sizes ranging from ~1.3-250 μm
	Electrical resistance particle sizing	Sizes ranging from ~0.6-12 μm and ~2.4-48 μm
	Light Microscopy	Sizes ranging from ~1-250 μm
	SEM	Sizes ranging from ~0.01-100 μm
Bulk refractive index	Goniometer	Index-matching immersion method

Closure between measured particle characteristics and optical measurements including backscattering is being tested with theoretical models to help understand and interpret backscattering sources.

A component of our approach is the development of a new microphotometric method for determining the backscattering properties of individual particles. This technique allows evaluation of backscatter from specific structural components of a particle, providing extraordinary insight into the causative agents of bulk backscattering.

Testing the detrital and mineral hypothesis

The detrital and mineral hypotheses is best tested in samples from natural waters, preferably case 1 type waters without significant terrigenous influence. The measurements listed in Table 1 are being made in the field and the presence of detrital material and inorganic material is being determined directly (e.g., PSD and SEM analyses) and indirectly (e.g., inversion of optical measurements). Backscattering budgets will be composed.

Discrete samples will also be sequentially filtered and the VSFs will be measured in the different size fractions with the goniometer. Samples will also be filtered in-situ with real-time instrumentation including the AC9 by attaching capsule filters to the intakes of the sensors. Isolating the scattering characteristics of individual size fractions, especially in the submicron range, will allow discrimination of the PSD component(s) thought to be responsible for backscattering under the detrital hypothesis.

Standard gravimetric determinations of TSM are also being made to determine relative and absolute concentrations of minerals in samples. Concentrations and isolated scattering characteristics of the mineral fraction will allow an assessment of the overall role of mineral particles in the oceanic backscattering process. Particles collected on filters for TSM analyses are also being subjected to SEM and elemental analysis with a mass spectrometer integrated in an SEM microscope. This analysis can directly determine the type and concentration of individual particles on the filters. Elemental analyses leave virtually no doubt as to mineral type and consequently the refractive index of that particle group (from published tables in CRC). An SEM image analysis method was developed to obtain PSDs from particles collected on a filter.

WORK COMPLETED

- A method was developed to determine the VSF of the colloidal fraction only in a whole sample using the goniometer system.

- A microphotometric method was developed to assess the backscattering properties of individual particles and internal structures.

- A method was developed to obtain PSDs from analysis of SEM images of particles collected on a filter.

- 5 lab experiments have been conducted to assess the complex particle hypothesis using cultures of the diatom *Chaetoceros socialis*, the dinoflagellate *Gyrodinium instriatum*, the diatom *Thalassiosira weissflogii*, the diatom *Chaetoceros teres*, and the diatom *Stephanopyxis turris*. The experiment with the latter organism was carried out through a full phytoplankton growth cycle.

- Parameters from Table 1 were collected on a cruise in the NW Atlantic in 10/ 2005.

• Parameters from Table 1 were collected on a cruise in Long Island Sound in 08/2006.

• Parameters from Table 1 collected from a cruise to the very clear waters of the South Pacific were analyzed to assess sources of backscattering. Particles collected from filtered samples were analyzed with SEM to determine PSDs and particle composition.

• A manuscript is in preparation for publication, entitled "The Contribution of Phytoplankton to Backscattering in Coastal Waters" with Twardowski/Sullivan leads.

• A manuscript is in preparation for publication, entitled "Sources of Backscattering in the Southeast Pacific" with Twardowski lead.

• A manuscript is in preparation for publication, entitled "Relative Influences of Spectral Absorption and Backscattering on Remote Sensing of Chlorophyll" with Dierssen, lead.

RESULTS

Previous measurements of phytoplankton scattering in the lab have critical errors. In culture work, we found that the contribution of the background particle population to scattering, particularly at large angles, is substantial. By gently filtering the phytoplankton particles out of the culture solutions with a 10 um nitex screen, we were able to quantify the optical properties of the background. The background particle population had dramatically different relative (e.g., backscattering ratio) and absolute scattering properties. To our knowledge, no previous study has considered the background solution in cultures.

Individual particle backscattering can be imaged with a new microphotometry technique. A new backscattering imaging method was developed to better assess where the backscattering from single particles was originating. At this time, the method is qualitative, but has proven highly valuable in helping us to better understand sources of particulate backscattering and develop a strategy for better addressing the hypotheses presented on the previous page. Results from this work have indicated that a hard coat is the primary source of backscattering for a phytoplankton cell. This was not necessarily the case for all cells, however (e.g., *Stephanopyxis*). And some phytoplankton are nearly invisible in the backscattering image (e.g., *Chaetoceros*). Additionally, the backscattering from detrital material, in particular the so-called Transparent Exo-Polymers (TEP), had far higher relative backscattering than previously expected based on analyses of the interstitial water content and subsequent refractive index of these particles (Twardowski et al. 2001).

Coastal phytoplankton have significantly higher backscattering than previously thought from Mie theory calculations. From an integrated analysis that included field measurements, lab measurements and theoretical considerations, we have demonstrated that the complex morphology of coastal phytoplankton substantially increases their relative backscattering. The higher backscattering is primarily due to their hard coats. This is clear from the new microscope laser images of single particle backscattering and is also supported by the consistency with expectations based on coated sphere Mie theory modeling (**Fig. 1**). Overall, a good rule of thumb backscattering ratio for coastal phytoplankton is ~0.5%, also supported by extensive field measurements. An approximate backscattering ratio

expected from homogeneous sphere Mie theory calculations is ~0.02-0.05% (more than an order of magnitude lower than actual) for coastal phytoplankton species (Stramski et al. 2000).

Scattering by colloidal material can be resolved without mechanically size fractionating a sample. Because of persistent particle contamination when trying to sequentially filter whole samples, a method was developed to measure the VSF of the colloidal fraction of a whole sample without any fractionation. Trying to first filter out the particles larger than colloids always resulted in substantial contamination of the colloidal fraction. The method is based on the principle that measuring scattering from as small a sample volume as possible with a very sensitive detector effectively excludes large particles (greater than a few um's) from the background colloidal scattering. A signal spike rejection algorithm allows the resolution of only this colloidal fraction, which has been verified with beads of colloidal and non-colloidal sizes. With a bench top goniometer, 1 s averaged measurements of scattering intensity are collected for each angle of the VSF until 5-10 consistent background measurements are recorded. Verification of calibration of this instrument is shown in **Fig. 2**.

Colloidal material accounted for ~50% of the backscattering in the Northwest Atlantic. The VSF of the colloidal fraction was measured in a sample from Northwest Atlantic surface water co-located with concurrent in situ ECO-VSF backscattering measurements (**Fig. 3**). The bench top goniometer was brought on the R/V Endeavor for these measurements. The backscattering from the colloidal fraction was about half that obtained with the real-time, in situ ECO-VSF sensor in the whole sample. This is the first time the contribution of colloids to backscattering in the ocean has ever been measured directly.

Conventional remote sensing chlorophyll algorithms generally work because of selective filtering of light (i.e., spectral absorption) rather than selective backscattering, except when chlorophyll concentrations exceed ~ 1 mg m^{-3}. Radiative transfer modeling of remote sensing reflectance from the IOPs has shown that phytoplankton backscattering does not need to dominate a water-leaving radiance signal for chlorophyll algorithms to work. The influence of backscattering from phytoplankton only starts to exceed the influence from spectral absorption when chlorophyll concentrations are greater than about 1 mg m^{-3}. These modeling results are consistent with experimental conclusions from the field and lab.

For additional results related to this project, refer to the annual reports of co-I's Jim Sullivan and Heidi Dierssen.

IMPACT/APPLICATIONS

Progress and results represent important steps toward understanding the sources of backscattering in the ocean. Naval applications requiring an understanding of the optical properties of water will benefit from this work. Since the optical properties of seawater are driven by the composition of suspended materials, we must understand this link to know how the underlying biogeochemical processes influence seawater optics. Applications directly influenced by seawater backscattering include lidar, laser line scanning systems, and remote sensing. Oceanographic research implications of this work include better inversion models for estimating the composition and concentration of suspended particles from optical sensors.

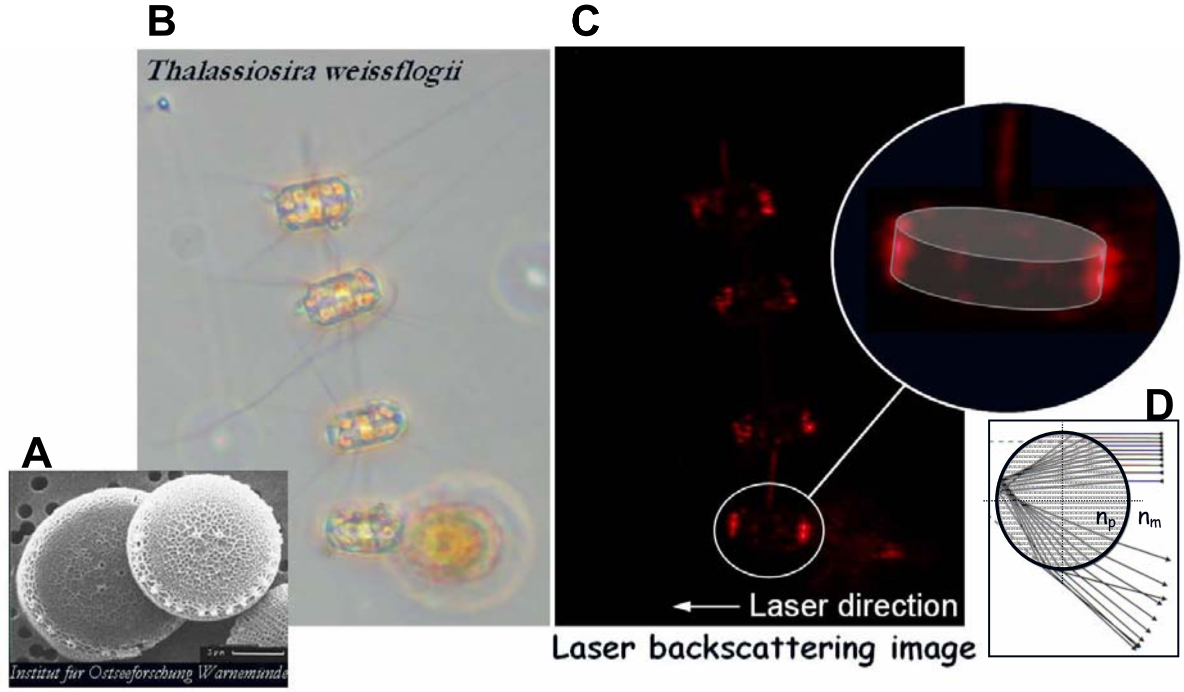

Figure 1. The importance of the "hard coat" in backscattering by coastal phytoplankton is illustrated by this microscopic backscattering image from a chain of Thalassiosira weissflogii cells. An SEM image (A) shows the heavily silicified hard coat (frustule) of the diatom cells. Images (B) and (C) are of the same field-of-view, with the (B) image obtained with conventional light transmission microscopy and the (C) image obtained from laser backscattering at ~140 degrees. Backscattering "hot spots" were observed at the outer edges of the frustule from ray focusing, analogous to the effect achieved with a focusing lens (D). The observed effect results from significant refraction of the incident rays that is only possible if there is a relatively high refractive index difference between the medium and the particle (or particle shell in this case). These images provide, for the first time, experimental microscopic evidence of the importance of complex structure in particle backscattering.

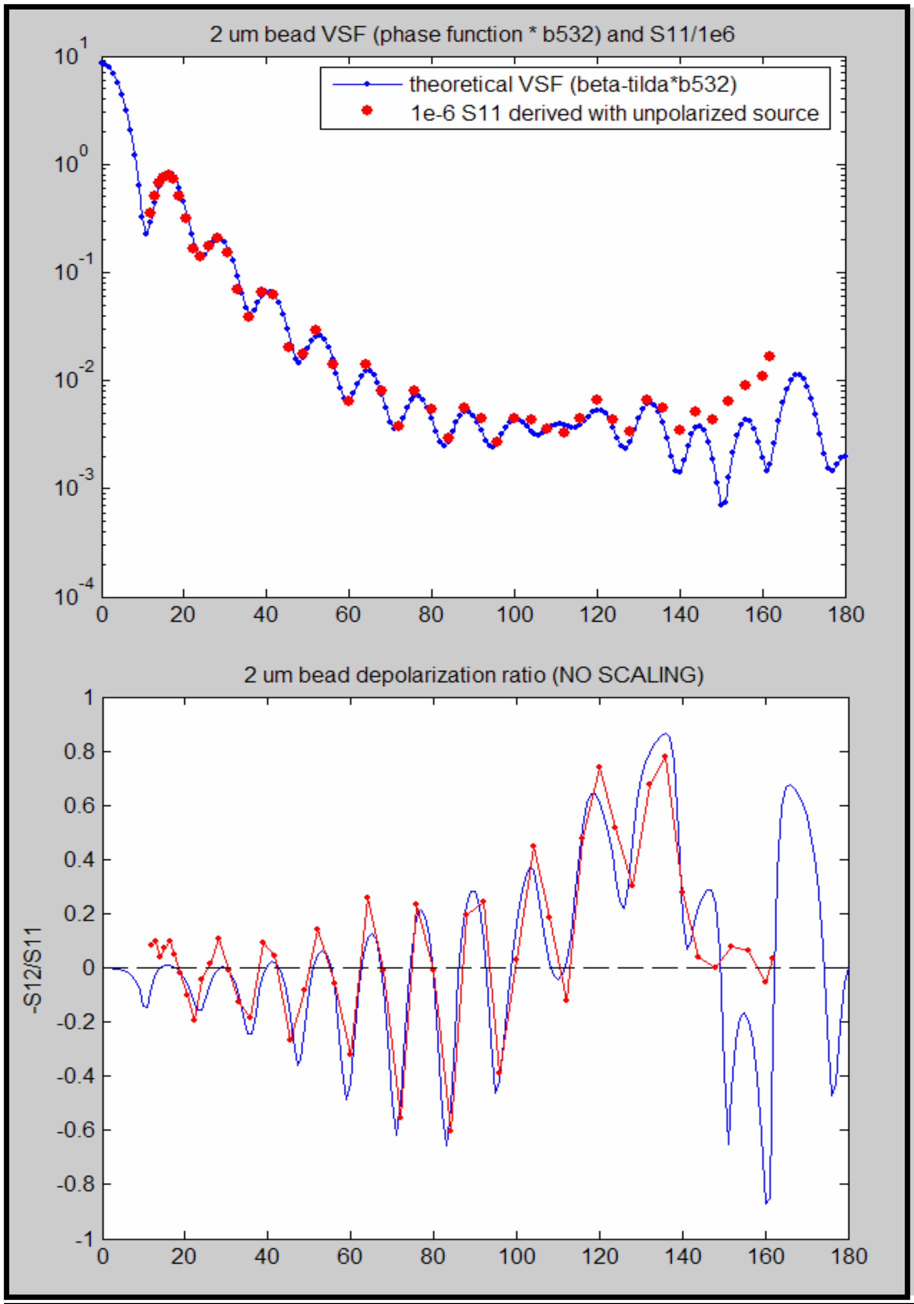

Figure 2. Validation of calibration of the bench top goniometer with 2 μm microspherical beads. Results agree well with theoretical expectations from Mie theory except at large angles (>~140 degrees) where system reflections begin to contaminate the signal.

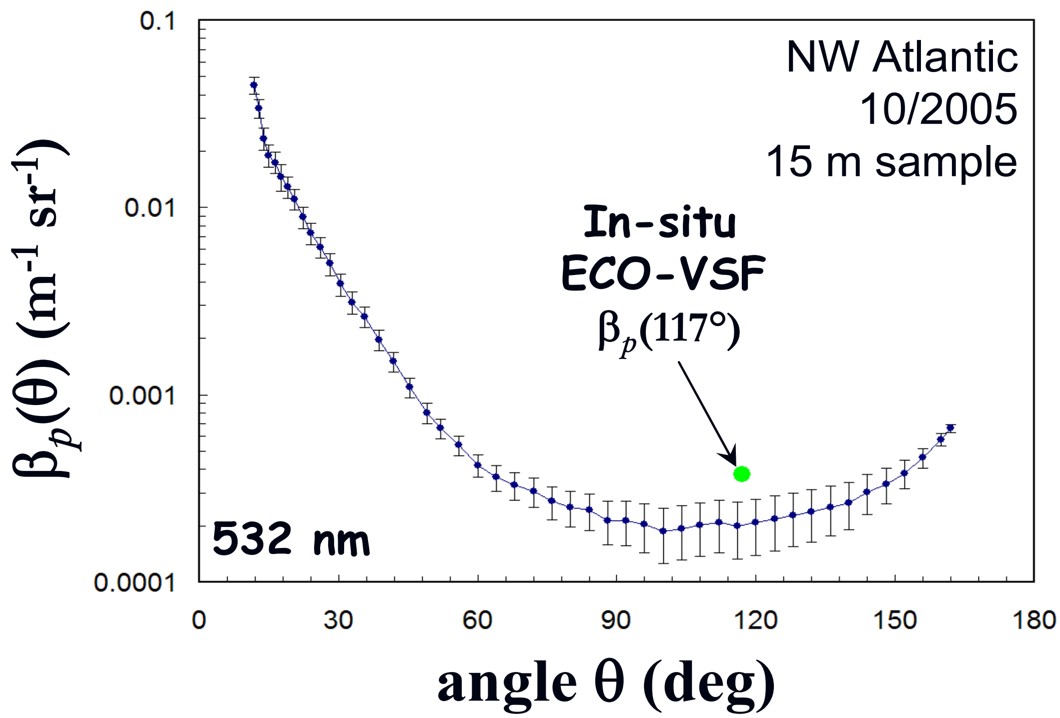

Figure 3. High-resolution VSF of the colloidal fraction of seawater collected from the NW Atlantic. The VSF was determined with a bench top goniometer using a new statistical large particle (>a few μm's) rejection algorithm. The green data point was collected with an in-situ ECO-VSF sensor and represents the volume scattering at 117 degrees by the total particulate fraction. Assuming scattering at 117 degrees is a reasonable proxy for integrated backscattering (Oishi 1990; Boss and Pegau 2001), the colloidal fraction comprised about half the particulate backscattering. These results verify experimentally for the first time that colloids represent a significant, if not dominant, portion of particulate backscattering.

TRANSITIONS

Sullivan, Twardowski et al. have completed an analysis of the hyperspectral temperature and salinity dependencies of pure water absorption and attenuation that is being made available to the community through WET Labs for ACS corrections. This work was published in 2006.

In 2005, Twardowski et al. published a chapter in Remote Sensing of Coastal Aquatic Environments entitled "In-water instrumentation and platforms for ocean color remote sensing applications." Twardowski has agreed to teach a short course on Observational Approaches in Ocean Optics at the next Ocean Optics conference in Montreal in 2006 where this work will serve as a backbone.

Three new methods were developed under this contract that we expect will be of value to the community. These methods are being prepared for publication.

Work on measuring and inverting VSFs in natural samples has been incorporated into conceptual sensor designs for two new R&D efforts at WET Labs. One device is a miniaturized total scattering device for AUVs and the second is a VSF device called the Multiple Angle Scattering and Optical Transmission (MASCOT) sensor. WET Labs plans to commercialize both sensors.

RELATED PROJECTS

This effort is related to several ongoing efforts to develop biogeochemical inversion techniques based on scattering measurements. Related projects include:

- resolving the optics and dynamics of bottom nepheloid layers with miniaturized sensors on gliders,

- developing miniaturized optical sensors for gliders and other compact platforms,

- developing a multi-angle volume scattering meter for resolving particle field VSFs in the ocean subsurface,

- developing novel harbor security monitoring capabilities with Chuck Trees and Jim Mueller,

- developing improved vicarious calibration and validation methods for ocean color satellite remote sensing, and

- developing tools for ocean observing systems in collaboration with Andrew Barnard, Percy Donaghay, and Jim Sullivan.

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PATENTS

Scattering attenuation meter (SAM), pending.

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